

**TRANSPORTATION FUEL CELL RESEARCH AT
ARGONNE NATIONAL LABORATORY***

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Introduction

Three major areas of transportation fuel cell research at Argonne National Laboratory (ANL) are discussed in this presentation. The first of these is fuel processor development for converting methanol and other liquid fuels to a hydrogen-rich fuel gas for use in the fuel cell stack. The second research area is fuel cell systems modeling: designing and analyzing polymer electrolyte fuel cell systems for use in light-duty vehicles. Lastly, this paper discusses the development of a facility at ANL for testing and characterizing the performance of fuel cells, fuel cell stacks, and complete systems.

Fuel Processor Development

We are developing a catalyzed partial-oxidation fuel reformer (APOR) for methanol and hydrocarbon fuels. At present, the only fuel cell-powered vehicles operating on a liquid fuel are the DOE/DOT/Georgetown University 30-ft transit buses. These buses have 50-kW phosphoric acid fuel cell systems that operate on methanol, converting it to hydrogen by steam reforming. However, the steam reformers are large (416 L, 14.7 ft³), heavy (266 kg, 586 lb), and slow to start (30 min or more). These characteristics obviously make such steam reformers unacceptable for light-duty vehicles.

The major factor limiting the performance of a steam reformer is the need for external heat input into the process gas. Therefore, they are designed primarily as heat exchangers. Their design is controlled by the limited practical heat fluxes obtainable in indirect heat transfer. A second performance-limiting factor is the conventional steam reforming catalyst for methanol, supported copper-zinc oxide, which must first be activated by reducing it with hydrogen before it can be used. Once the catalyst is reduced, it must not come in contact with air, which would reoxidize it and render it ineffective. The reduced form of the catalyst is also highly susceptible to sintering (and consequent deactivation) during use if temperatures much above 250°C are encountered.

The Partnership for a New Generation of Vehicles (PNGV) has established tentative targets for automotive fuel cell systems. For a fuel processor in a 50-kW system the target weight and volume are 50 kg and 100 L, respectively. The fuel processor should start up in 1 min or less and it should have a transient response time of 10 s or less. The target cost is \$500 or less.

These requirements can be met by catalytic partial-oxidation reforming. Such reforming operates as a combination of oxidation reforming, steam reforming, methanol decomposition, and the water-gas shift reaction. Methanol oxidation is highly exothermic, while the others are either

endothermic or nearly thermally neutral. Thus, the net energy release (or absorption) of catalytic partial-oxidation reforming is varied primarily by changing the oxygen-to-methanol ratio (and secondarily by varying the water-to-methanol ratio) in the reformer feed. Varying these parameters also affects the hydrogen content of the reformat. For maximum reforming efficiency, such a reformer is best operated close to the thermoneutral point. A slight excess of oxygen (air) is needed to provide for the sensible heating of the reformat above the ambient temperature and the heat losses from the reformer. A hydrogen concentration of 50% or higher (on a dry basis) is obtainable from methanol under such conditions.

The APOR is simple in design and construction. The methanol and water are injected as a fine spray into a flowing air stream. An electrically heated nichrome-wire coil serves to vaporize a small fraction of the liquid methanol. This vapor readily undergoes the reforming reactions on the surface of the catalyst. The copper-zinc oxide catalyst is used in the thermally rugged oxide form that does not need to be isolated from air between uses. The bench-scale APOR yields a hydrogen production rate equivalent to 6 kWe. The weight and volume of the catalyst in the APOR are 1.8 kg and 1.8 L, respectively, well within the PNGV target values mentioned above. The APOR starts up rapidly, reaching operating temperatures in 2 min or less. At steady-state, the reformat contains (on a dry basis) approximately 50% H_2 , 20% CO_2 , and less than 1% CO (the balance is N_2). This composition is maintained even after step changes in the fuel processing rates, showing the excellent transient capability of the reformer.

The APOR has been demonstrated for methanol. It is now being considered for other liquid fuels, such as gasoline and diesel fuel. For this purpose, catalysts are being identified that can convert the individual classes of hydrocarbons in gasoline (such as paraffins, olefins, aromatics, and naphthenes) to hydrogen by partial-oxidation reforming. The thermoneutral point for the reforming of octane, for example, is at an oxygen-to-octane molar ratio of 2.9; at this operating point a hydrogen concentration of 50% is theoretically possible. Preliminary data using novel catalysts indicate that such reforming is possible in the temperature range of 450–550°C.

Fuel Cell Systems Modeling

Analytical models have been developed to study the dynamic response of steam-reformed, methanol-fueled, polymer electrolyte fuel cell systems. For the dynamic analyses, attention was focused primarily on the heat transfer effects that are likely to limit rapid response of such systems. Depending on the thermal mass, the heat exchangers and the steam reformer can have time constants on the order of several seconds to several minutes. The characteristic time constants of pressure/density variations arising from flow rate fluctuations are on the order of milliseconds. In vehicular applications, the response time of the turbo-machinery, which is determined by its rotational inertia, is generally on the order of seconds or less. Thus, the controlling time constants for the dynamic response of fuel cell systems are those related to the heat exchange required in steam reformers. They are being studied at length to determine design and operational characteristics that would yield the best dynamic performance.

The dynamic methanol steam-reformer model has been used to examine the methanol conversion efficiency and thermal performance during cold starts. Response times were determined to achieve 50–100% of the steady-state methanol conversion in a generic shell-and-tube-type steam reformer for two different catalyst tube diameters. The resulting thermal performance was analyzed

in terms of the approach to steady-state temperatures, the possibility of catalyst overheating, and the penalty in system efficiency incurred during the start-up.

For the complete fuel cell power system, various turn-down scenarios were simulated by varying the relative rates of change of fuel cell loading and system flows. It was found that, depending on the rates of cell loading changes relative to flow rate changes, overheating of the catalyst can occur due to excess heat transfer in the reformer preheater. This overheating can be controlled by an additional water quench between the preheater and the catalyst bed, but only if the flow rate change is sufficiently fast relative to the load changes. For example, in one such power turn-down scenario, the flow rates of the fuel gas and air were turned down in 1 s, while maintaining the fuel utilization (u_f) constant at 85%. For a 50% reduction in power level, the simulation showed that the reformer catalyst overheated in less than 10 s. This overheating was alleviated by injecting water into the process gas just ahead of the reformer. However, if the fuel gas flow rate is not decreased in concert with the decrease in fuel cell power (i.e., u_f is initially permitted to decrease to 25%, then gradually increased to 85%), combustion of the excess hydrogen in the spent fuel gas at the reformer burner rapidly leads to unacceptably high reformer catalyst temperatures.

Fuel Cell Test Facility

To satisfy the need for an independent source of fuel cell performance data, evaluation, and system validation, ANL is constructing a test facility for fuel cell, stacks, and systems ranging in size from 10 kW to 60 kW. This facility will use standard conditions and test procedures to determine the performance, operational characteristics, and durability of such systems. Extensive post-test analysis capabilities and technical expertise in fuel cell and energy storage systems are also available at ANL. This facility will be available for use by fuel cell developers, programmatic (e.g., PNGV) sponsors, as well as the automobile manufacturers.

This test facility will make use of the existing Electrochemical Analysis and Diagnostic Laboratory, a premier facility for evaluating batteries for transportation applications. This laboratory is equipped to test batteries on complex power profiles (representative of various vehicle driving schedules) and has proven control and data acquisition systems that can be readily adapted to fuel cell testing.

In the near term, the test facility will be used to test 10-kW stacks (the building blocks) to 60-kW systems (power output in the range of practical light-duty vehicles). Characterization of the performance and operation will be based on fuel type, temperatures, pressures, fuel and oxidant flow rates, humidification levels, and the test power profiles. Performance degradation over time and over cyclic operation will be determined. In the longer term, evaluation of balance-of-plant components, including fuel reformers, air compressors/expanders, and energy storage devices, will be conducted as well.

A major operational component of the test facility is the gas management system. This system controls both the compressed air and fuel supply gas streams to provide the test pressure, temperature, humidity, and flow rate of the fuel and oxidant gases. In addition, it will monitor the conditions of the exhaust gas streams. Any of these conditions may be used to control the operation of the test fuel cell, stack, or system, as desired.